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## ARSENIC CONTAMINATION OF WATER-SOIL-CROP SYSTEM IN AN INDUSTRIAL AREA OF BANGLADESH

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### Abstract

The arsenic (As) concentrations of irrigation water, soils and vegetables were investigated in an agricultural section of Gazipur industrial area in Bangladesh, where industrial wastewater is mixed with irrigation water. The results showed that the mean As concentration of the irrigation water (0.16-0.62 mg L<sup>-1</sup>) exceeded the permissible limit for agricultural purposes recommended by the Food and Agriculture Organization (0.10 mg L<sup>-1</sup>). The mean As concentration of soils (6.48-9.75 mg kg<sup>-1</sup>) did not exceed the tolerable limit for agricultural soils recommended by the FAO (20.0 mg kg<sup>-1</sup>). While, the As concentration of the respective vegetables in average varied from 0.63 to 1.07 mg kg<sup>-1</sup> dry weight, and the highest As concentration in average was observed in taro root (1.26-2.31 mg kg<sup>-1</sup>), followed by helencha leaf (1.85-2.02 mg kg<sup>-1</sup>). The average As concentration of root vegetables (1.84 mg kg<sup>-1</sup>) exceeded the permissible limit of 1.0 mg kg<sup>-1</sup> suggested by the Food and Agriculture Organization, while that of leafy (0.77 mg kg<sup>-1</sup>) and fruit vegetables (0.14 mg kg<sup>-1</sup>) did not exceed the limit. Soils irrigated with As containing water that holds much amount of As showed a positive correlation with the vegetable As concentration. A decrease in the As concentration from soil to vegetable was found in root, leafy and fruit vegetables. However, the vegetables were not safe for human consumption. Some countermeasures are, therefore, necessary to reduce the vegetable As concentration.

Key words: Arsenic, Industrial Wastewater, Vegetables, Permissible Limit

## **Introduction**

Heavy metal contaminations of surface and groundwater, agricultural soils and crops are the major concern for public health and ecosystem preservation. Recently, contamination of groundwater by arsenic (As) has attracted much attention compared to that by other heavy metals (Schwarzenbach et al., 2006, Krabbenhoft et al., 2013). The contamination is thought to be of anthropogenic (human-caused) origin. Surface water may also be contaminated by As. The As contamination of surface water occurs in different ways such as by the weathering of minerals (Mailloux et al., 2009, Foley and Ayuso, 2008) and by the anthropogenic activities that includes discharge of industrial waste water, applications of sewage sludge and waste water in agriculture, and mining activities. The exposure of As in human is caused by ingesting groundwater and crops contaminated by As. The exposure leads to the problems with cardiovascular diseases, premalignant skin lesions, conjunctivitis, etc. (Guha-Mazumder et al., 2000). The World Health Organization (WHO) treated the problems as “the largest poisoning of a population in history” (Smith et al., 2000).

In Bangladesh, many garment factories dispose their wastes into nearby rivers, canals and lakes without any treatment, and this disposal deteriorates surrounding environment and aquatic ecosystems. The waste water, containing As and other heavy metals, are used for irrigation in adjacent agricultural land. Various industries like textile, dyeing of plastics, metal fabrications, paints, lather, tanning, semiconductor goods, etc. are the major source of As and other heavy metals in the wastewater of Bangladesh (Ahmed et al., 2012). Continuous irrigation with As containing wastewater is quite responsible for increased As concentration in soil (Das et al., 2004). Crops grown in that soil can easily uptake As and accumulate it in their edible parts (Bhumla and Keefer, 1994).

The As concentration in the crops is raised by the uptake of As by crops from the contaminated soils, and the rise in the concentration is remarkably observed in vegetables and edible crops (Larsen et al., 1992). Irrigation with As-contaminated water causes As accumulation in crops like rice, wheat, fruit plants and vegetables (Roychowdhury et al., 2002). The observed As concentration in different crops varied widely from 0.007 to 7.50 mg kg<sup>-1</sup> (Mandal and Suzuki, 2002, Roychowdhury et al., 2002, Dahal et al., 2008).

There is a growing concern on the As contamination of crops in the industrial areas in Bangladesh. However, the actual situation of As contamination of crops in relation with As contamination of irrigation water and soils has not been clarified yet. Therefore, the present study aimed to clarify the magnitude of the As concentration of irrigation water, soils and crops, and the relationship of As concentration between irrigation water, soils and crops targeting an intensive industrial area in Bangladesh.

## **Materials and methods**

### **Site description and geology**

The study area is the main industrial areas in Gazipur District that are comprised of complex relief and soils developed over the Madhupur Clay, the Madhupur Tract. According to US soil taxonomy, Gazipur region belongs to Entisols mostly. The landscapes are level upland, closely or broadly dissected terraces associated with either shallow or broad, deep valleys and the drainage pattern is clearly dendritic (Banglapedia, 2003). Eleven general soil types are distributed but deep red brown and shallow red brown terrace soils and acid basin clays, which are nutrient poor, are dominant in the area.

Gazipur District has a tropical climate where summers are much rainier than the winters. This climate is considered to be Aw (Tropical wet and dry or savanna climate) according to the Köppen-Geiger climate classification. The average temperature in Gazipur District is 25.8°C. The annual average rainfall is 2036 mm. It is large industrial area comprised of a good number of local and foreign industries like garments, fabric printing and dyeing, food processing, textiles, electric cables, pharmaceutical, chemical, glass industry, ceramic industry etc. Paddy, wheat, potato, jute, mustard, ginger, turmeric, vegetables are the main crops of this region (Banglapedia, 2003). Different cropping system is practiced by the farmers for growing crops and vegetables. In this industrial area, crops are irrigated usually by the water of nearby canals and the industrial waste water as and when required.

### **Irrigation water, soils, and vegetables sampling**

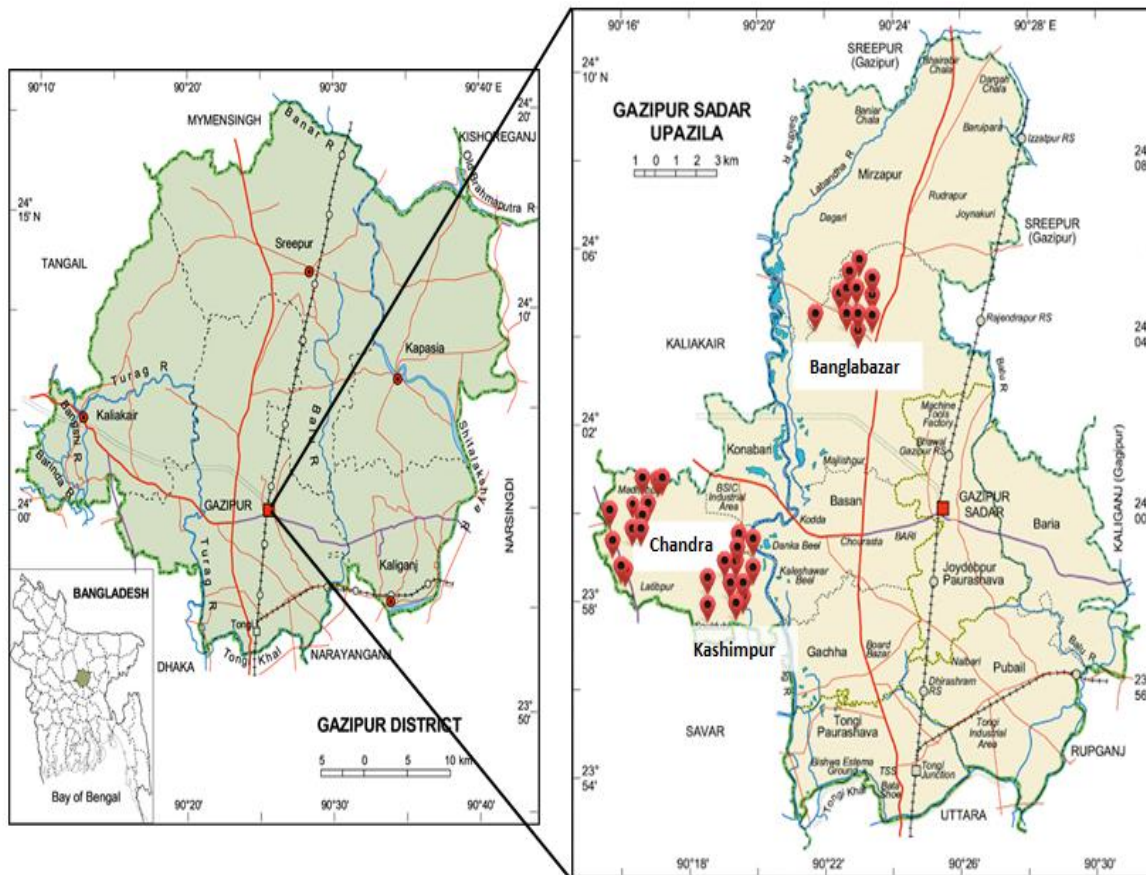
Three industrial areas viz. Banglabazar, Kashimpur and Chandra located in Gazipur District were targeted for the sampling (Figure 1). The sampling was done in June-July of 2015. For collecting irrigation water, 12 points were selected from each area, i.e. 36 points from all areas. Here, industrial wastewaters from various industries were mixed with irrigation water. Five hundred mL of irrigation water was taken from irrigation canal and stored it in sterilized air-tight plastic bottle and then it was moved to the laboratory. The sample was filtered through Whatman no. 42 paper filter and then a drop of HNO<sub>3</sub> (65%) was added to it to make pH<2 for preservation. The sample was kept in a 20 mL cleaned acid-washed plastic bottle and stored it in a refrigerator at 4°C until analysis.

Soils were collected from 12 points in each area as same as that of irrigation water. A total of 36 samples were collected. Soil was taken from the top soil layer (0-15 cm) of vegetable bed using an auger and the sample was packaged into transparent zipped lock plastic bag and then transferred to the laboratory. The soils were air-dried at room temperature for 7 days and grounded with mortar and pestle and sieved through a 2 mm sieve.

Vegetables collected were those planted in an agricultural season of Kharif 1. There are three agricultural seasons, namely Kharif 1 (March-July), Kharif 2 (July-October) and Rabi (October-March) in Bangladesh (Krishi Diary, 2015). The sampling period was the same with those of irrigation water and soils. The 5 kinds commonly grown vegetables viz. taro (*Colocasia esculenta*), brinjal (*Solanum melongena*), sponge gourd (*Luffa acutangula*), kang kong (*Ipomoea reptans*) and Helencha (*Enhydra fluctuans*) were collected from 9 vegetable beds for each area. Namely, a total of 27 samples were collected from all areas. The vegetable sampling spots were the same spots with those of soil sampling. Vegetables were washed with tap water followed by deionized water. After then, vegetables were separated into root, stem, leaf and fruit, and these were air-dried and dried using drying oven at 60<sup>0</sup>C for 48 hours for completely drying and milled it into powder with a grinder.

### **Sample analysis**

The irrigation water samples were diluted with pure water mixed with 2% (v/v) HNO<sub>3</sub>. For soil analysis, 1g of each sample was digested with concentrated HNO<sub>3</sub> and 30% H<sub>2</sub>O<sub>2</sub> at 95°C (USEPA 3050B method; USEPA, 1996) until a clear digested solution was obtained. The digested solutions (approximately 5 mL) were filtered through a paper filter (No. 5B, 125 mm, Advantec Tokyo, Japan), and diluted to 100 mL with pure water. For the analysis of vegetables, 0.5 g each sample was digested with concentrated HNO<sub>3</sub> and 30% H<sub>2</sub>O<sub>2</sub> at 120-130°C for 14-16 hours (UWLAB, 2005). The digested solution was filtered with a paper filter (No. 5B, 125 mm, Advantec Tokyo, Japan) and diluted to 50 ml. The As concentrations of irrigation water and digested solutions of soils and vegetables (with following dilutions) were analyzed by inductively coupled plasma mass spectrometry (ICP-MS; Agilent 7500 ce, Agilent technologies) at Kyushu University, Japan.



**Figure 1: Location of the sampling points in Gazipur District, Bangladesh.**

## Results and discussion

### The As concentrations of irrigation water

The As concentration of irrigation water varied from 0.03 to 1.58 mg L<sup>-1</sup> in all areas (Table 1). The As concentrations in Banglabazar, Kashimpur, and Chandra in average were 0.62, 0.20 and 0.16 mg L<sup>-1</sup>, respectively, which are many fold higher than the FAO permissible limit of 0.10 mg L<sup>-1</sup> for agricultural purposes (FAO, 1985). There are various factories including textile mills, dyeing, plastics, metal fabrications, paints, etc. in the areas, and these factories are thought to be responsible for the high As concentration.

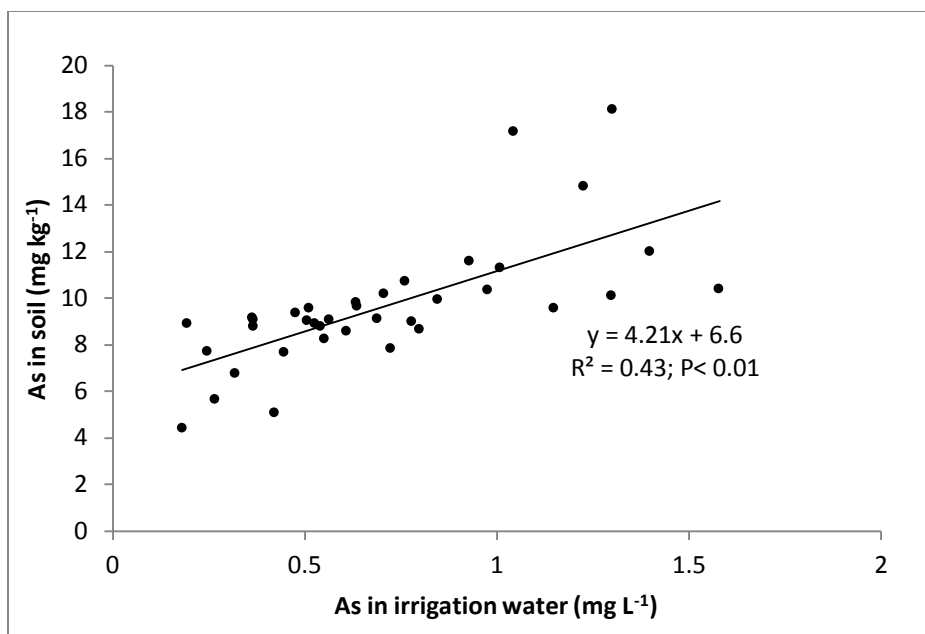
**Table 1: The As concentrations in irrigation water (mg L<sup>-1</sup>), soils (mg kg<sup>-1</sup>) and vegetables (mg kg<sup>-1</sup>) in the three areas in Gazipur District, Bangladesh**

Material	Area	Permissible limit <sup>a</sup>	Mean ± SD	Range
Irrigation water (n=12x3)	Banglabazar	0.10	0.62±0.28	0.25-1.58
	Kashimpur		0.20±0.15	0.06-0.78
	Chandra		0.16±0.13	0.03-0.61
Soils (n=12x3)	Banglabazar	20	9.75±2.91	5.01-18.11
	Kashimpur		8.39±1.77	4.43-12.23
	Chandra		6.48±1.03	4.56-9.21
Vegetables (n=9x3)	Banglabazar	1	1.07±0.67	0.17-2.31
	Kashimpur		0.63±0.07	0.05-2.03
	Chandra		0.69±0.51	0.07-1.91

<sup>a</sup> FAO standard, 1985

#### **The As concentrations of soils**

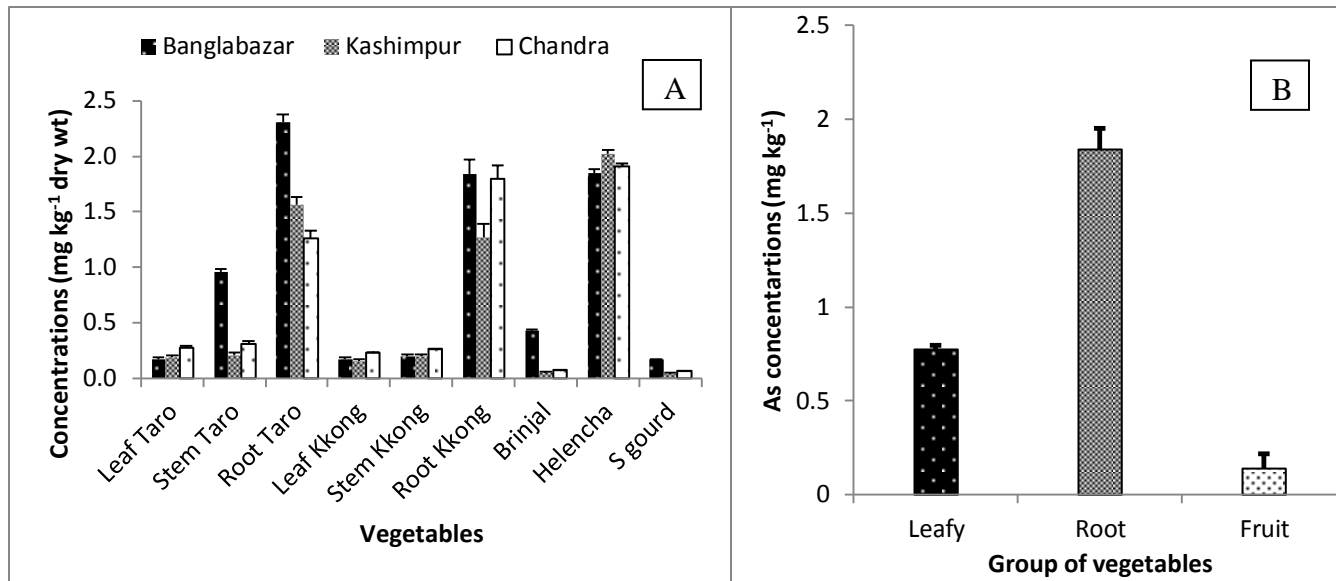
The As concentrations of soils in average were 9.75, 8.39 and 6.48 mg kg<sup>-1</sup> in Banglabazar, Kashimpur, and Chandra, respectively (Table 1), which satisfied the permissible limit for agricultural soils of 20 mg kg<sup>-1</sup> recommended by Food and Agriculture Organization (FAO, 1985). Relationship of As concentration between irrigation water and soils was depicted in Figure 2. According to Figure 2, a positive correlation was observed in the As concentration between irrigation water and soils ( $p < 0.01$ ). If an As contaminated irrigation water is used for a long term, the As would be accumulated in soils.



**Figure 2: Relationship of As concentration between irrigation water and soils.**

### **The As concentrations of vegetables**

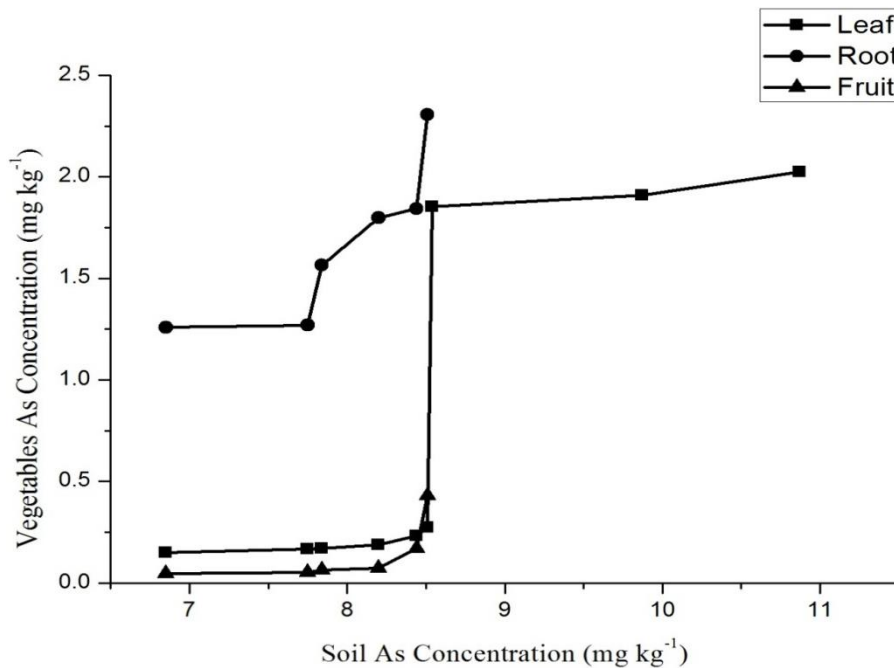
Figure 3 shows the As concentrations of vegetables according to the division of area, and kind and category of vegetables. According to Figure 3A, the As concentration of taro ranged from 0.16 to 2.31 mg kg<sup>-1</sup>. That of kang kong ranged from 0.15–1.84 mg kg<sup>-1</sup>. That of helencha ranged from 1.85 to 2.02 mg kg<sup>-1</sup>. The concentrations of brinjal and sponge gourd ranged from 0.07 - 0.43 mg kg<sup>-1</sup> and from 0.06 to 0.17 mg kg<sup>-1</sup>, respectively (Figure 3A). According to the grouping of leafy, root and fruit vegetables, the As concentration was in the order of root vegetables (1.84 mg kg<sup>-1</sup>)>leafy vegetables (0.77 mg kg<sup>-1</sup>)> fruit vegetables (0.14 mg kg<sup>-1</sup>) (Figure 3B). Among them only root vegetables exceed the permissible limit of 1.0 mg kg<sup>-1</sup> (FAO, 1985) compared to leafy and fruit vegetables. In previous study Aracil et al. (2001) found that the As concentration of edible beans (*Phaseolus vulgaris*) was below the FAO permissible limit. Root vegetables taro bears a short underground stem called a corm, where the plant stores starch produced by the leaves. It is up to 30 cm long and 15 cm in diameter. Whereas kang kong bear roots at the nodes and grows on the moist soil. These underground portions of root vegetables were supposed to contain more As than leaves and fruits.



**Figure 3: The As Concentration ( $\pm$  standard error of mean) of vegetables in averages.**

In general, the As in vegetable is thought to be sourced from soils (Huang et al., 2006). Most of the agricultural soils of the study area were irrigated with the water from nearby canals. The water was mixed with industrial waste water, therefore, the industrial waste water is probably the source of As. The relationship of As concentration between soils and vegetables in the study area is shown in Figure 4. According to Figure 4, vegetable As concentration increased with the increase in soil As concentration except for fruit vegetable, however, the increase rate and the concentration differed with vegetables. When the soil As concentrations were 6.85- 8.51 mg kg<sup>-1</sup>, the As concentration was high (1.26-2.31 mg kg<sup>-1</sup>) in root vegetable, while the concentration was low (<0.3 mg kg<sup>-1</sup>) in leafy and fruit vegetables. When the soil As concentrations were higher than 8.5 mg kg<sup>-1</sup>, the As concentration of leafy vegetable (helencha) became high with 1.85-2.02 mg kg<sup>-1</sup>. Helencha is water loving annual herb, grows in swampy grounds of the study area. For growing this vegetables much amount of irrigation water was applied which thought to be the main reason of the high As concentration. From the results, all of root and a part of leafy vegetables are found to be unsuitable for human consumption.





**Figure 4: Relationship between soil and plant As concentration.**

### Conclusions

The As concentrations in irrigation water and root vegetables in Gazipur District exceeded the permissible limit for agricultural purpose, while the soil As concentration satisfied this limit. A positive correlation of As concentration was found between irrigation water and cultivable soil. The rise of As concentration were also found from soil to root and leafy vegetables. All roots and a part of leafy vegetables are found to be unsuitable for human consumption.

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